THE ROCKY FLATS NUCLEAR WEAPONS PLANT DOSE RECONSTRUCTION AND RISK CHARACTERIZATION PROJECT

PHASE II: TOXICITY ASSESSMENT AND RISK CHARACTERIZATION

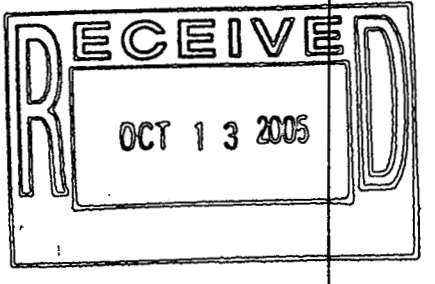
Technical Memorandum: EVALUATION OF BACKGROUND CONCENTRATIONS OF PLUTONIUM IN SOILS AROUND THE ROCKY FLATS PLANT

Prepared for the Colorado Department of Health,
Disease Control and Environmental Epidemiology,
Rocky Flats Health Studies,
in partial fulfillment of Contract No. 100APPR CODE 391

by

Radiological Assessments Corporation
417 Till Road
Neeses, South Carolina 29107
(803) 536-4883

September 1994



DRAFT FOR COMMENT

1/14

Technical Memorandum

EVALUATION OF BACKGROUND CONCENTRATIONS OF PLUTONIUM IN SOILS AROUND THE ROCKY FLATS PLANT

Duane W. Schmidt
Health Physics Applications
Darnestown, Maryland 20878

September 1994

SUMMARY

The evaluation of background levels of plutonium in the environment around the Rocky Flats Plant is important to the determination of the impacts of historical releases from the plant. Since essentially all plutonium in the environment is due to human activities, there is no "natural" background for plutonium. For our work, we define background plutonium to be plutonium in the environment from sources other than the Rocky Flats Plant.

The major aims of the work described in this memorandum are to:

- Gain an understanding of the important factors that must be considered in assessing measurements of plutonium in soils, for background determinations and for areas contaminated by emissions from the Rocky Flats Plant.
- Provide general perspective on background levels of plutonium in soils around the Rocky Flats Plant.

In this technical memorandum we discuss the sources of background environmental plutonium, and indicate which contribute to the background around the Rocky Flats Plant. We then describe important characteristics of environmental plutonium in soils that should be considered in evaluations of measurements of plutonium in soils. Finally we compile measurement data on background levels of plutonium in soils, both around the Rocky Flats Plant and around the United States.

The primary source of background plutonium in the Rocky Flats area is global fallout from the atmospheric testing of nuclear weapons. Of the alpha radiation-emitting isotopes of plutonium (the alpha-emitting isotopes are of most concern for this Project), the primary constituents of this global fallout are plutonium-239 and plutonium-240. A secondary source of background plutonium is global fallout from the atmospheric burnup of a satellite, which contributed about 1% of the total background (alpha-emitting) plutonium activity in the northern hemisphere. Plutonium from the satellite was essentially all plutonium-238. Regional fallout from activities at the Nevada Test Site may also have contributed small amounts to the plutonium background.

The evaluation of measured background plutonium levels in soil can be a difficult task. In such evaluations, there are several important factors that must be considered.

- The depth of soil sampled can be a key factor, and must be appropriate to the goals of the evaluation. Samples taken from surface soils (to a depth of 1 cm or so) are generally not representative of the total quantity of plutonium that has been deposited in the soil. Quantitative comparisons between studies using widely different sample depths will usually be difficult and unwise.
- The time of sample collection (especially the year) may also be important, because the total amount of plutonium in the environment increased over time in the 1950s and 1960s, reaching almost constant levels in the 1970s.
- The analysis methods should be determined, if possible. Many results reported as "plutonium-239" are actually plutonium-239 plus plutonium-240.
- Isotopic ratios (such as the ratio of plutonium-240 to plutonium-239) can be more powerful than the simpler analyses of total plutonium-239 plus plutonium-240. Such ratios can be used to ascertain the sources of the measured plutonium (for example, from weapons fallout versus from the Rocky Flats Plant).

Two types of results for plutonium in soils have been reported: (1) concentrations of plutonium per unit mass of soil (mass concentrations), with units such as Bq kg^{-1} or pCi g^{-1}; and (2) total quantity of plutonium that has been deposited on and remains in the soil, per unit soil surface area (deposition), with units such as Bq m^{-2} or mCi km^{-2}. Based on a number of historical studies, we make preliminary estimates of the range of background plutonium in soils around the Rocky Flats Plant.

- The background mass concentration of plutonium-239 plus plutonium-240 in soils around the Rocky Flats Plant is probably in the range 0.3–4 Bq kg^{-1} (0.008–0.1 pCi g^{-1}).
- The background deposition of plutonium-239 plus plutonium-240 around the Rocky Flats Plant is probably in the range 40–100 Bq m^{-2} (1.1–2.7 mCi km^{-2}).
- These background plutonium levels in soil around the Rocky Flats Plant are generally within background ranges observed in other locations in the United States, but tend to be on the higher side of these ranges.
- Levels of background plutonium in soils can vary considerably by location, with latitude being an important factor accounting for differences between locations.
- We consider these ranges rough estimates, to be used for general perspective, because there may be data that have not been evaluated. In addition, Colorado State University is currently engaged in additional sampling and analysis to determine background plutonium in soil; results from their study are expected shortly.

INTRODUCTION

The purposes of this technical memorandum are to describe important factors that must be considered in assessing measurements of plutonium (Pu) in soils, for background determinations and for areas contaminated by emissions from the Rocky Flats Plant, and to provide general perspective on background levels of plutonium in soils around the Rocky

Flats Plant (RFP). This information will be useful for anticipated evaluations of historical measurements, to be developed in Task 4 of the Project. The primary focus of this material is on Pu in soils. For the work of Task 4, information about other radionuclides and media will be developed by the RAC Team.

The radioactivity sources of concern for this Project are radioactive emissions from the RFP. Thus, we define background environmental Pu to be Pu in the environment that is due to sources other than the RFP, even though such sources also arise from human activities. In this memorandum, we describe sources of environmental Pu around the RFP, characteristics of environmental Pu that may be important to consider in evaluating soil sample results, measured background levels of Pu in soils around the RFP, and measured background levels of Pu in soils across the United States.

A variety of units have been used in the literature for expressing concentrations of Pu in soils and other media. We try to avoid confusion by converting concentrations to SI units (Bq kg^{-1} or Bq m^{-2}) when we wish to compare different sets of values. We also often provide the units used in the original source document. In some cases, especially when we are mostly interested in the relative results from a single study, only the units from the original source document are provided. Two types of results for plutonium in soils have been reported: (1) concentrations of plutonium per unit mass of soil (mass concentrations), with units such as Bq kg^{-1} or pCi g^{-1}; and (2) total quantity of plutonium that has been deposited on and remains in the soil, per unit soil surface area (deposition), with units such as Bq m^{-2} or mCi km^{-2}. We note that it is generally inadvisable to try to convert results between mass concentrations and values of total deposition, without knowledge of the depth distribution of plutonium.

SOURCES OF ENVIRONMENTAL PLUTONIUM

Essentially all plutonium in the environment is artificial, arising from activities of humans. The most important sources of environmental Pu are described in this section, with the emphasis on sources that may significantly contribute to the background Pu around the RFP. Harley (1979) provides a review of sources of Pu, and the following material is primarily obtained from that report.

Sources of environmental Pu can be categorized as global sources, that have distributed Pu around the world, and local sources, that distribute Pu on a much smaller spatial scale. Global sources include atmospheric nuclear weapons testing and the burnup of a satellite in the atmosphere. Local sources include releases from nuclear facilities and accidental releases.

Global Fallout from Nuclear Weapons Testing

Atmospheric nuclear weapons testing is the largest source of Pu in the environment (Harley 1979). Radionuclides formed in nuclear weapons tests are summarized by Holleman et al. (1987). Weapons-grade Pu is composed primarily (weight-basis) of ^{239}Pu, but also includes ^{238}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. In nuclear explosions, both fission and fusion weapons release Pu; this is derived from unused Pu (that does not fission) and from neutron

capture reactions, which create the majority of the higher-mass isotopes ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{243}Pu. The quantities of ^{242}Pu and ^{243}Pu produced are very small. Very large quantities of ^{241}Pu are produced. However, ^{241}Pu decays primarily by weak beta emissions, and its radiological impacts are much less significant than for the primary alpha-emitting Pu isotopes, ^{238}Pu, ^{239}Pu, and ^{240}Pu. Other heavy-element radionuclides are also released in nuclear weapons tests, including ^{237}U, ^{239}Np, and ^{241}Am. Uranium-237 and ^{239}Np have relatively short half-lives, of about 7 days and 2 days, respectively and would not persist in the environment. Americium-241 builds up in the environment as a result of ^{241}Pu decay, and thus is present in the environment in significant quantities relative to the primary alpha-emitting Pu isotopes. However, at this point we concentrate on the primary alpha-emitting Pu isotopes ^{238}Pu, ^{239}Pu, and ^{240}Pu.

Eisenbud (1987) summarizes the history of nuclear weapons testing. Atmospheric testing of nuclear weapons started in New Mexico in July 1945. The majority of tests were performed by the United States, the USSR, the United Kingdom, France, and China. Most of the atmospheric tests were performed in the 1950s and the early 1960s, before the signing of an atmospheric nuclear weapons test ban agreement in 1963 by the United States, the USSR, and the United Kingdom. The announced atmospheric tests (Eisenbud 1987) are summarized in Table 1, with estimated yields given in units of megatons of TNT which would produce an equivalent explosive yield.

Table 1. Summary of Announced Atmospheric Nuclear Weapons Tests [a]

Country	Period	Number of tests	Estimated total yield (megatons)
United States	1945–1962	193	139
USSR	1949–1962	142	358
United Kingdom	1952–1953	21	17
France	1960–1974	45	12
China	1964–1980	22	21
Total		423	547

[a] Adapted from Eisenbud (1987).

Holleman et al. (1987) provides a summary of the atmospheric transport of fallout from nuclear weapons testing. Because nuclear explosions create extremely high temperatures, a fireball is formed after the explosion. The expanding fireball can rise many kilometers, carrying debris from the explosion with it, and reaches greater altitude for higher yield weapons. At low yields, from about 10 to about 200 kilotons, all of the debris remains in the troposphere. At high yields (1 to 2 megatons), 90 to 99% of the debris reaches the stratosphere. Transport of the material is dependent on the height at which the fireball initially injects the debris.

Particles formed in the nuclear explosion can be transported long distances by winds. Material reaching the stratosphere is transported around the globe. Deposition of material from the atmosphere occurs by dry deposition or by wet deposition. To a great extent, air

masses of the northern and southern hemispheres remain separated, though limited exchange between hemispheres does occur.

Three additional features are relevant to global fallout in the area around the RFP. Mountains alter wind currents, resulting in a downward mixing of higher altitude air, which then increases the ground-level air concentrations of fallout on the lee side of the mountains. Also, high mountain passes and the lee side of mountains generally receive more precipitation than surrounding areas, increasing the wet deposition of global fallout. For meteorological reasons, material from the stratosphere is transferred into the troposphere primarily over the middle latitudes (about 40° to 50° latitude) (UNSCEAR 1993). Thus, the greatest amounts of fallout from large tests, which reach the stratosphere, are eventually distributed in the middle latitudes, with lesser amounts toward the poles and the equator (UNSCEAR 1993). The RFP is in the middle latitudes, at about 40° north latitude. Distribution of fallout from lower-yield tests is dependent on the location of the explosion.

The small particles of debris from atmospheric nuclear weapons tests may remain in the atmosphere for quite some time. For material that reaches the stratosphere, residence times are generally determined to be about 2–4 years. Thus, tests that inject debris into the stratosphere generally do not produce the highest ground-level fallout concentrations until about 2 years after the explosions.

Global Fallout from SNAP 9A Burnup

This global source of Pu is described by. In April 1964 a Transit Navigational Satellite was launched from California. Part of the payload was an auxiliary power generator (called SNAP 9A), which contained 17 kCi (6.3×10^{14} Bq) ^{238}Pu (Harley 1979). The rocket system failed, and the satellite reentered the atmosphere in the southern hemisphere, burning up at about 50 km altitude upon reentry. Essentially all of the Pu activity was ^{238}Pu. The first arrival in the northern hemisphere of plutonium-238 fallout from the satellite burnup was measured in early 1966 in Italy. This source of Pu contributes a small amount to the background total Pu in the RFP area (see Table 2 for general comparison).

Localized Sources

There are a number of sources of localized Pu in the environment, including both accidental releases and releases from nuclear processing facilities (Harley 1979). The Nevada Test Site (NTS), in southern Nevada, was used for test detonations of small nuclear weapons up through 1961, safety tests in which the high explosives in nuclear weapons were detonated (with Pu in the tested device), Plowshare explosions (using nuclear explosions for peaceful purposes), and accidental venting of underground weapons tests. There was a considerable amount of unfissioned Pu distributed from these tests. Material from the NTS was distributed at least as far away as Salt Lake City, Utah (about 600 km from the NTS), and may have contributed small amounts to the Pu deposition in Colorado.

There were two incidents in which the chemical explosives in nuclear weapons exploded, following crashes of U.S. military aircraft. The first occurred in Palomares, Spain, in 1966, and the second in Thule, Greenland, in 1968. Both resulted in local dispersion of Pu from the weapons.

A number of the U.S. atmospheric nuclear weapons tests were performed in the Pacific Ocean, at Bikini and Enewetak Atolls. Because large quantities of Pu were produced in these tests, there was significant Pu deposited in the local area around the tests.

France, India, and the United Kingdom have also released significant quantities of Pu to oceans, in effluents and as packaged waste for disposal (Harley 1979). Essentially all of this material has remained in the oceans.

Several of the Department of Energy weapons plants in the U.S. process Pu, and releases have occurred from some of them (Harley 1979). At the Mound facility in Ohio, a liquid release of about 10 Ci (4×10^{11} Bq) ^{238}Pu occurred in 1969, from a break in a low-level waste pipeline. At the Los Alamos National Laboratory, in New Mexico, about 2 Ci (8×10^{10} Bq) Pu has been released to canyon waste disposal sites. Harley (1979) indicates that releases have also occurred from the Hanford site, in Washington, and the Savannah River Plant, in South Carolina. Eisenbud (1987) indicates that a Pu release occurred at the Oak Ridge National Laboratory, in Tennessee. All of these releases resulted only in localized deposition of Pu.

And, as discussed in other documents of the Rocky Flats Dose Reconstruction Project, there have been routine and accidental releases of Pu from the RFP.

Summary of Sources of Plutonium in the Rocky Flats Area

Of the sources of environmental Pu, significant contributors to Pu around the RFP are global fallout from atmospheric weapons testing, fallout from the burnup of the SNAP 9A unit, releases from the RFP, and potentially regional fallout from activities at the Nevada Test Site. Pu from global fallout and the SNAP burnup are thought to be the primary contributors to the background Pu around the RFP. Table 2 summarizes the quantities of Pu measured in soils in 1970, at a number of locations in the northern and southern hemispheres, that were due to weapons tests and the SNAP 9A burnup (Harley 1979). These values are presented to give a rough indication of global trends in fallout Pu levels. The values in this table for the northern hemisphere are more pertinent to the RFP. The background Pu is primarily due to ^{239,240}Pu from weapons fallout.

Table 2. Plutonium Inventories (kCi) Measured in Soils in 1970 [a]

Location	Weapons ^{239,240}Pu	Weapons ^{238}Pu	SNAP ^{238}Pu
Northern hemisphere	253 ± 33	6.1 ± 0.8	3.1 ± 0.8
Southern hemisphere	67 ± 14	1.6 ± 0.3	10.3 ± 2.1
Total	320 ± 36	7.7 ± 0.9	13.4 ± 2.2

[a] Source: Harley 1979.

CHARACTERISTICS OF ENVIRONMENTAL PLUTONIUM

In this section we discuss some characteristics of environmental Pu, related both to background Pu and to Pu from the RFP. Some of these characteristics are particularly important for consideration in evaluations of soil concentrations of Pu.

Measurement of Plutonium

Holleman et al. (1987) provides a summary of analytical techniques commonly used for Pu analyses in environmental samples. The most common analytical technique uses alpha spectroscopy, a procedure which segregates alpha particle radiations by energy. This allows the differentiation of $^{239,240}Pu$ from ^{238}Pu. However, since the alpha energies of ^{239}Pu and ^{240}Pu are quite similar, these two isotopes cannot be separately determined with alpha spectroscopy. Sometimes, values reported as ^{239}Pu are actually measurements of $^{239,240}Pu$. If differentiation of ^{239}Pu and ^{240}Pu is required, mass spectrometry can be used, though it is not as readily applied to field samples. Alpha spectroscopy results are generally reported in radiological units, while mass spectrometry results are usually reported in mass units, or as atom ratios relative to ^{239}Pu.

Sample preparation for alpha spectroscopy counting first requires extraction of the Pu from the sample media. This is usually performed with concentrated acid, or filter samples may be wet ashed. Chemical separations are then performed, and the Pu is plated out on a probe (or planchet) for counting in an alpha spectrometer. Plutonium-236 or ^{242}Pu can be added as a tracer at the start of sample preparation to help measure the chemical recovery (Krey and Hardy 1970 and Holleman et al. 1987).

Isotopic Composition of Global Fallout and Rocky Flats Plutonium

The Pu processed at the RFP is weapons-grade Pu, consisting primarily of ^{239}Pu. Plutonium from atmospheric nuclear weapons tests is weapons-grade Pu that has undergone (partial) fission and neutron capture reactions in the nuclear explosion. Because of these reactions, the relative abundance of the various Pu isotopes is altered in the exploded material.

Krey and Krajewski measured the isotopic Pu composition of a soil sample thought to contain Pu essentially only from RFP releases, and a sample from New York thought to contain only global fallout Pu. Isotopic ratios, relative to ^{239}Pu, are compared in Table 3.

Table 3. Mass Isotopic Ratios, Relative to ^{239}Pu, for Soil Samples Contaminated by Rocky Flats Plutonium or by Global Fallout Plutonium, from Krey and Krajewski (1972)

Sample	$^{238}Pu/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$	$^{242}Pu/^{239}Pu$
RFP Pu	$7.15 \times 10^{-5} \pm 9\%$	$5.10 \times 10^{-2} \pm 0.19\%$	$2.31 \times 10^{-3} \pm 0.42\%$	$1.43 \times 10^{-4} \pm 1.7\%$
Global fallout	$1.35 \times 10^{-4} \pm 5\%$	$1.80 \times 10^{-1} \pm 0.24\%$	$7.76 \times 10^{-3} \pm 0.5\%$	$3.89 \times 10^{-3} \pm 0.59\%$

Other sources have reported slightly different isotopic ratios for global fallout Pu. Krey (1976) reported measurements for two samples from New York, with an average ratio $^{240}Pu/^{239}Pu$ of 0.163 ± 0.008. Bennett (1978) reported measured Pu isotopic mass ratios for stratospheric air samples for 1959–1970. Average measured ratios were 0.18 for $^{240}Pu/^{239}Pu$; 0.0138 and 0.0118 for $^{241}Pu/^{239}Pu$ for 1963–1967 and for other years, respectively; and 0.0034 for $^{242}Pu/^{239}Pu$. While there may be slight differences in isotopic ratios in samples of global fallout Pu, the isotopic ratios for RFP Pu are significantly different than those for global fallout material. These significant differences can and have

been used to differentiate between RFP Pu and global fallout Pu, and to determine which source dominates in a particular soil sample (some results are presented later in this memorandum). It appears that the most frequently used ratio is $^{240}Pu/^{239}Pu$. One reason for the use of $^{240}Pu/^{239}Pu$ is that the higher abundance of ^{240}Pu, compared to other isotopes, results in smaller uncertainties.

Temporal Trends in Global Fallout

In using background concentrations of Pu in the environment for comparisons with concentrations on and around the RFP, it can be important to recognize the temporal trends (changes with time) in the global fallout background. The major temporal trend in fallout Pu concentrations is due to the temporal distribution of the weapons tests that were the source of the fallout Pu. Bennett (1978) summarizes the estimated explosive yields of nuclear weapons tests, and this information is shown in Table 4. The cumulative yield is plotted later, in Figure 2.

Table 4. Summary of Total Explosive Yields (Megatons) from Atmospheric Nuclear Tests

Period	Total explosive yield	Cumulative yield
1945–1951	0.75	0.75
1952–1954	60.52	61.27
1955–1956	30.79	92.06
1957–1958	81.39	173.45
1960–1961	122.43	295.88
1962	217.40	513.28
1964–1970	21.23	534.51
1971–1974	6.46	540.97
1976–1978	4.16	545.13

We examine the temporal trends in fallout Pu by reviewing modeling predictions performed by Bennett (1978). We do not rely on these predictions for explicit, quantitative uses; they are presented to give an appreciation of the general trends.

Bennett (1978) used the information about the weapons testing with an atmospheric transport model to predict fallout concentrations of Pu and Am in surface air in the middle latitudes of the northern hemisphere. Table 5 shows the predicted air concentrations of $^{239,240}Pu$, and Figure 1 is a plot of these concentrations. The air concentrations of Pu from global fallout vary considerably. It is important to consider this temporal trend of air concentrations when measured concentrations around the RFP are compared to background concentrations. Because of the seasonal changes in global winds, there are also seasonal trends in fallout air concentrations (Holleman et al. 1987). For short-term air concentration measurements, these seasonal trends should be considered. Since our focus here is toward soil samples, the seasonal trends are not examined in more detail.

Table 5. Predicted Surface Air Concentrations of $^{239,240}Pu$ (fCi m^{-3}) from Global Fallout from Nuclear Weapons Testing [a]

Year	Concentration	Year	Concentration	Year	Concentration
1950	0	1961	0.089	1972	0.032
1951	0.0001	1962	0.54	1973	0.021
1952	0.0022	1963	1.18	1974	0.028
1953	0.031	1964	0.58	1975	0.017
1954	0.097	1965	0.25	1976	0.0083
1955	0.16	1966	0.11	1977	0.044
1956	0.14	1967	0.054	1978	0.018
1957	0.19	1968	0.042	1979	0.0071
1958	0.25	1969	0.056	1980	0.0028
1959	0.33	1970	0.062		
1960	0.11	1971	0.066		

[a] Source: Bennett 1978.

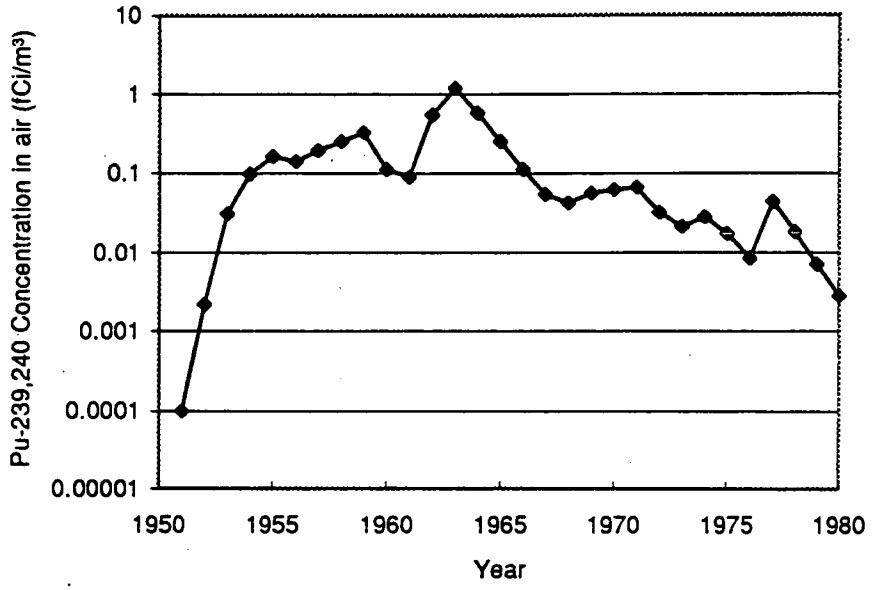


Figure 1. Predicted surface air concentrations of $^{239,240}Pu$ (fCi m^{-3}) from global fallout from nuclear weapons testing.

Bennett (1978) also used the atmospheric transport model to predict deposition rates and cumulative deposition of Pu and Am in the New York region. These predictions are shown in Table 6. The predicted cumulative deposition of $^{239,240}Pu$ is plotted in Figure 2, along with the cumulative yield of the weapons tests for comparison. The predicted cumulative deposition follows the same general shape as the cumulative yield, but after a lag time. The temporal trend in the cumulative deposition of fallout Pu should be considered

when comparing RFP-influenced soil sample results with background results. This trend can be especially important when comparing samples taken at different times. We note that the predicted cumulative deposition of Pu from nuclear weapons fallout reaches about 90% of its predicted maximum value in 1968, and reaches 95% of maximum in 1971 (this is relevant to the background soil samples discussed later in this memorandum). Although these predicted depositions are for New York, they should also be relevant to the RFP area, since both locations are in the middle latitudes.

Table 6. Predicted Deposition Rate and Cumulative Deposition of ^{239,240}Pu in the New York Area due to Global Fallout from Nuclear Weapons Testing [a]

Year	Deposition rate (mCi km^{-2} y^{-1})	Cumulative deposition (mCi km^{-2})	Year	Deposition rate (mCi km^{-2} y^{-1})	Cumulative deposition (mCi km^{-2})
1950	0	0	1968	0.021	2.00
1951	0.00006	0.00039	1969	0.047	2.04
1952	0.0012	0.0013	1970	0.031	2.07
1953	0.017	0.019	1971	0.029	2.10
1954	0.054	0.072	1972	0.023	2.13
1955	0.091	0.16	1973	0.017	2.14
1956	0.077	0.24	1974	0.018	2.16
1957	0.11	0.35	1975	0.012	2.17
1958	0.14	0.49	1976	0.0075	2.18
1959	0.19	0.67	1977	0.024	2.20
1960	0.061	0.73	1978	0.0098	2.21
1961	0.049	0.78	1979	0.0039	2.22
1962	0.30	1.08	1980	0.0016	2.22
1963	0.44	1.52	1981	0.00061	2.22
1964	0.26	1.78	1982	0.00022	2.22
1965	0.11	1.89	1983	0.00011	2.22
1966	0.046	1.93	1984	0.00006	2.22
1967	0.042	1.98			

[a] Source: Bennett 1978.

Depth Distribution of Plutonium in Soils

As seen later in this memorandum, many of the background soil samples analyzed for Pu content were taken from surface soils of 0–1 cm depth or less, or from relatively thick samples of 0–20 cm or 0–10 cm depth. It is important to consider the depth distribution of Pu in soils, when samples of such different depths are evaluated.

A few historical studies around the RFP have investigated the depth distribution of Pu in soils, and we briefly discuss them here. Krey and Hardy (1970) investigated the depth distribution at eight sampling locations around the RFP, from just next to the plant to distances up to about 40 miles from the site. Based on these samples, between 9 and 61% of the ^{239,240}Pu from the surface to 20 cm depth was below 5 cm depth. Two other sample locations, in New York City and Waynesville, Ohio, showed results within this same range.

From limited sampling at finer depth resolution, it was concluded that less than 1% of the Pu occurred below a depth of 13 cm.

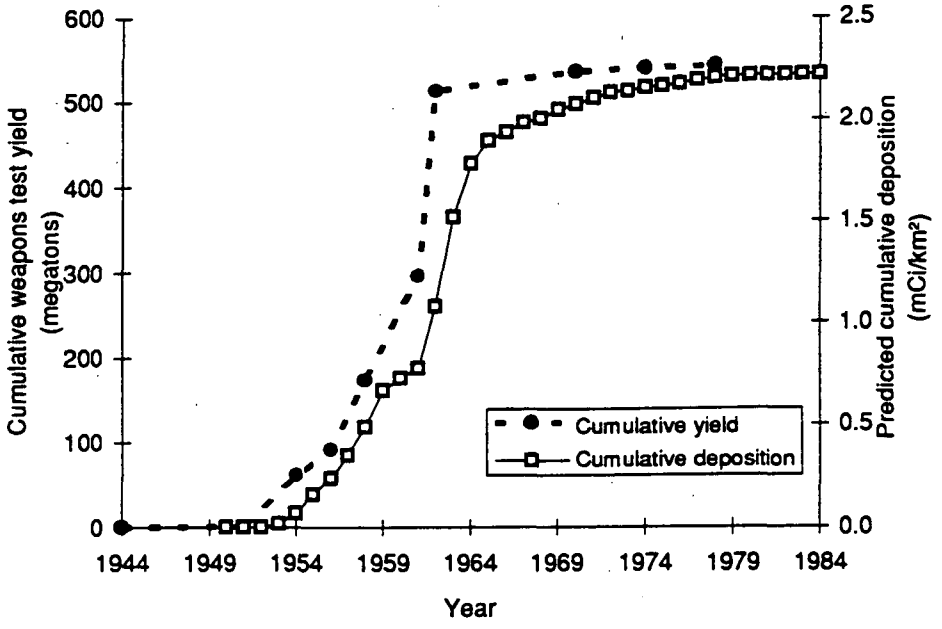


Figure 2. Predicted cumulative deposition of Pu in the New York area due to global fallout from nuclear weapons testing. For comparison, the cumulative explosive yield of atmospheric weapons tests is also plotted.

Whicker et al. (1974) performed preliminary investigations of the inventories of ^{239}Pu in several compartments of the terrestrial ecosystem near the RFP. Two locations were studied; one a contaminated area very close to the former oil barrel storage pad, and the other a relatively less contaminated area (the control) south of the main plant (though not representative of background Pu). Results from this study indicated that for both the contaminated study area and the control area about 40% of the Pu in soil occurred below 3 cm depth.

Little and Whicker (1978) performed more detailed investigations of the depth distributions of Pu in soil near the RFP (as well as particle size distributions). This work utilized the same sampling areas as that of Whicker et al. (1974). Samples were taken in 3 cm-thick samples to a depth of 21 cm. Four replicate samples from each layer were obtained from ten locations in the contaminated area and from five locations in the control area. We have used the average concentrations reported by Little and Whicker, for the less than 2 mm size particles, to calculate the fractional contributions of each depth interval to the total Pu content. For these calculations, we have assumed a constant soil density for all layers of soil. Results are shown in Table 7. Since the soil densities for each sample layer were not available, the calculated relative Pu activities are not exact. (Also, due to rounding the percentages do not add to exactly 100%.) Qualitatively, it is clear that although much of

the Pu activity occurs in the top 3 cm of soil, layers to as deep as 21 cm are significant contributors to the total soil column Pu.

Table 7. Estimated Depth Distributions of ^{239,240}Pu in Soils from Two Areas Near the Rocky Flats Plant, from Little and Whicker (1978)

	Average relative Pu activity in each sample layer (percent of total)						
Area	0–3 cm	3–6 cm	6–9 cm	9–12 cm	12–15 cm	15–18 cm	18–21 cm
Contaminated	58	11	18	4.6	5.8	2.3	0.82
Control	70.	6.1	3.4	3.2	4.4	7.1	6.1

In 1989 Webb et al. (1993) also studied the contaminated area previously studied by Whicker et al. (1974). The report by Webb et al. also presented additional results from sampling in 1972–1974. The inventory of ^{239}Pu was measured for soil layers 0–3 cm and 3–21 cm. It was concluded that for both periods about 50% of the soil inventory was in each of these layers. However, the soil concentration in the 0–3 cm layer was significantly less in 1989 than in 1972–1974. To account for this, Webb et al. made the preliminary speculation that about 2.75 cm of the surface soil had been lost from the study area by soil erosion over the fifteen years between the two studies.

The studies described here have resulted in different depth distributions of Pu in the soil column. However, it is clear from the results that significant quantities of Pu have migrated to depths of at least 10–20 cm. Whenever possible, these results should be considered in the evaluations of soil Pu concentration measurements around the RFP. In particular, we note that it is not reasonable to use measurements of the surface soil concentrations of Pu (to depths of 1 cm or so) to estimate the total inventory (or total deposition) of Pu in the soil. This is the reason that later in this memorandum, we do not try to directly compare background surface soil Pu concentrations (expressed as Bq kg^{-1}, or similar) to background soil Pu inventories (expressed in Bq m^{-2}, or similar).

BACKGROUND PLUTONIUM IN SOIL NEAR THE ROCKY FLATS PLANT

In this section we describe results from studies around the Rocky Flats Plant that represent background concentrations of Pu in soil. The fire at the Rocky Flats Plant in 1969 caused an increased interest in monitoring soil concentrations of Pu around the Plant. A number of soil monitoring studies around the Plant were performed or begun in late 1969 and in the early 1970s. Studies were performed by the National Center for Atmospheric Research (NCAR) for the Colorado Committee for Environmental Information (CCEI), the Colorado Department of Health (CDH), and the Health and Safety Laboratory (HASL) of the Atomic Energy Commission.

The NCAR and CDH obtained surface samples (0.3 and 1 cm depths) and reported results as mass concentrations, while HASL obtained samples to 10 and 20 cm depths and reported results as total deposition (per unit area). The shallow depths of the NCAR and CDH samples mean that not all of the Pu in the soil column was sampled. It is not

reasonable to convert the mass concentration results of NCAR and CDH to total deposition values, and thus the NCAR and CDH results can not be directly compared with HASL results.

The first study was performed by NCAR for CCEI in late 1969 and early 1970. Results were reported first by CCEI (CCEI 1970), with additional results given in the later report by NCAR (Poet and Martell 1972). This study sampled soils at 35 locations around the Plant and in the Denver area, and three locations on the eastern slope of the Rocky Mountains that were thought to contain Pu only from nuclear weapons fallout. For this study, surface soil samples were taken to a depth of 1 cm. The background sampling locations are shown in Figure 3. Results from the background locations are provided in Table 8. Analysis errors (standard deviations) are included to provide general perspective on the analytical precision. Results were given in units dpm g^{-1} (dpm means disintegrations per minute), and we have converted to Bq kg^{-1} in Table 8.

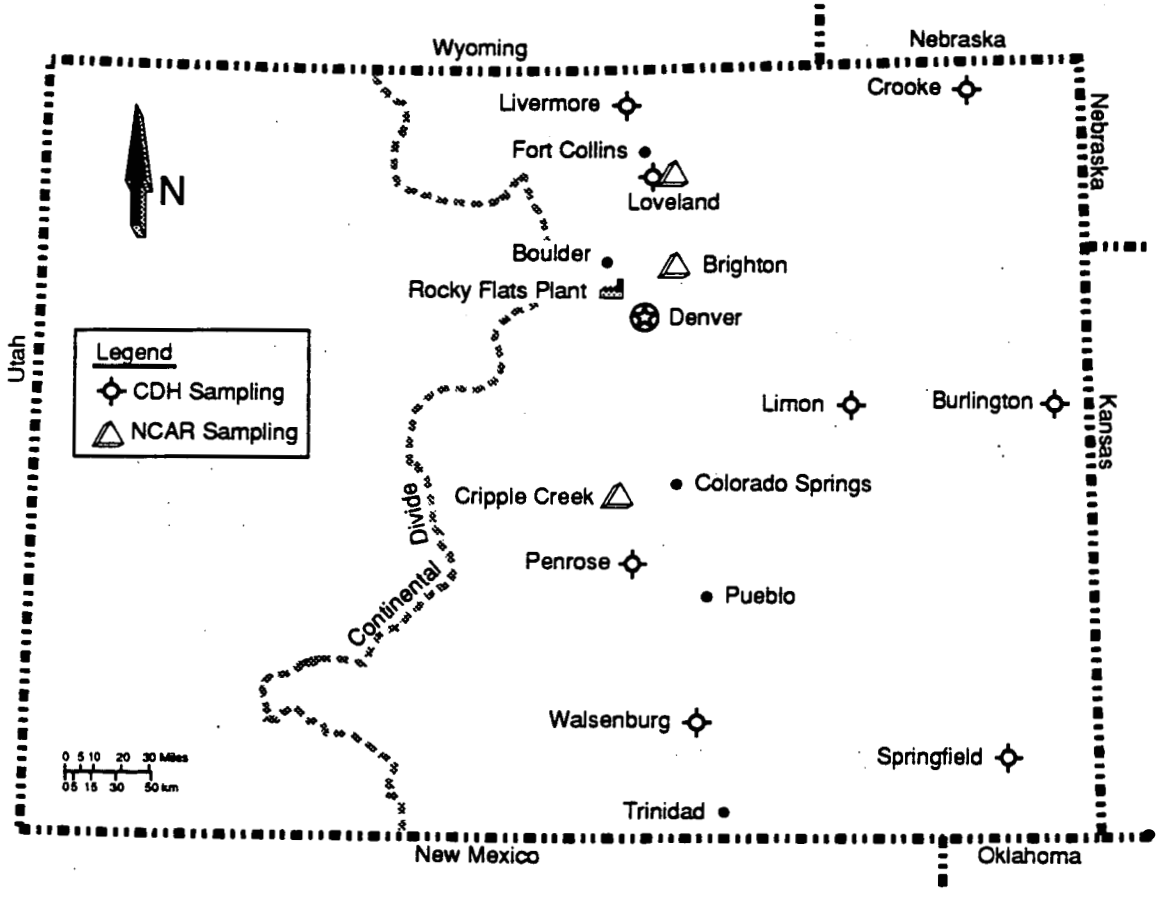


Figure 3. Background sample locations, in Colorado, of the NCAR and CDH studies.

Soil sampling around the RFP was also performed by the CDH. Results of monitoring for 1970–1977 are presented by CDH (1977 and 1990). Samples were generally collected from 13 sectors near the RFP and eight remote sites in Colorado each year, though in some years

not all the sites were sampled. The remote site locations are shown in Figure 3. The CDH used its own method to obtain samples for 1975–1988. This method includes taking 25 individual samples, to a depth of 0.3 cm (one-eighth inch), at each site and then compositing to form a single sample for analysis. Surface soil is sampled. The sampling procedures used for years prior to 1975 were not detailed. Results from the background locations for 1976 and 1977 are provided in Table 9. Analysis errors (2 σ) are also shown, to provide general perspective on the analytical precision. Results for 1970–1986 are summarized in Table 10, though no results for these background locations were available for 1974, 1979, and 1981–1985. Results were given in units dpm g^{-1}, which we have converted to Bq kg^{-1} in Table 9 and Table 10.

Table 8. Background (Fallout) Concentrations of Plutonium in Soil Measured Around the Rocky Flats Plant in 1969–1970

	Concentration of $^{239,240}Pu$ [a]			
	dpm g^{-1}		Bq kg^{-1}	
Location	Value	Std dev.[b]	Value	Std dev.[b]
Loveland	0.047	0.013	0.78	0.22
Loveland	0.056	0.025	0.93	0.42
Loveland	0.045	0.008	0.75	0.13
Loveland	0.026	0.006	0.43	0.1
Loveland	0.043	0.005	0.72	0.08
Brighton	0.093	0.009	1.6	0.15
Cripple Creek	0.140	0.027	2.3	0.45
Cripple Creek	0.052	0.012	0.87	0.20
Cripple Creek	0.117	0.015	2.0	0.25
Weighted average[c]	0.034	0.005	0.57	0.083

[a] The source document (Poet and Martell 1972) gives results in units dpm g^{-1}.
[b] "Std. dev." means standard deviation.
[c] Weighted average as taken from source document (Poet and Martell 1972). Weighting is inversely proportional to the percent error represented by one standard deviation.

Studies of Pu in soil around the RFP by the Health and Safety Laboratory (HASL) of the U.S. Atomic Energy Commission were initiated in early 1970. These studies did not separately select background sampling locations, as done in the studies described above. Instead, sample locations were chosen at increasing distances from the RFP, and analytical techniques were generally employed to estimate background concentrations. The first study by the HASL is reported by Krey and Hardy (1970). Samples were collected in February 1970 from 33 sites around the RFP, to distances of about 40 miles, and primarily in easterly

directions from the site. Figure 4 shows the numbered locations (1–33), except for some of those close to the plant. Samples were collected to a depth of 8 inches (20 cm). At some locations depth profile information was obtained by collecting samples in incremental layers to a total depth of 8 inches. Based on limited depth profile information, Krey and Hardy concluded that less than 1% of the total Pu in soil was deeper than 13 cm. Results from this study were expressed in units mCi km^{-2} total deposited Pu, based on the assumption that the measured Pu (to depth of 20 cm) was the total deposited Pu. Since the studies described earlier in this section used shallow sample depths, their results can not be reasonably compared to results of these HASL studies.

Table 9. Background (Fallout) Concentrations of Plutonium-239,240 in Soil (Bq kg^{-1}) Measured in Colorado in 1976 and 1977 [a]

	1976		1977	
Location	Value	Counting error (2 σ)	Value	Counting error (2 σ)
Loveland	[b]		0.3	0.3
Livermore	0.3	0.3	<0.3	
Crooke	1.3 [c]	0.2	0.7	0.3
Burlington	0.3	0.3	1.2	0.3
Limon	0.7	0.3	0.7	0.3
Springfield	0.3	0.3	0.3	0.3
Walsenburg	[b]		0.7	0.7
Penrose	1.5	0.7	0.7	0.3

[a] Values were reported in units dpm g^{-1} in the source document (CDH 1977).
[b] No sample was taken at this location in 1976.
[c] Average of two samples.

Table 10. Background (Fallout) Concentrations of Plutonium-239,240 in Soil (Bq kg^{-1}) Measured in Colorado in 1970–1986 [a]

Location	1970	1971	1972	1973	1975	1976	1977	1978	1980	1986
Loveland	1.8	1.7	[b]	2.0	[b]	[b]	0.3	<0.7	[b]	<0.7
Livermore	0.7	<0.7	1.2	0.7	[b]	0.3	<0.3	0.7	[b]	<0.7
Crooke	0.7	2.2	1.8	0.8	[b]	1.3	0.7	1.2	[b]	<0.3
Burlington	1.5	1.8	1.2	0.8	[b]	0.3	1.2	<0.7	0.7	<0.3
Limon	2.2	1.0	1.2	1.0	[b]	0.7	0.7	<0.7	0.7	<1.2
Springfield	0.7	1.5	2.0	<0.7	0.7	0.3	0.3	4.5	0.7	<1.5
Walsenburg	1.8	1.2	1.8	0.8	0.3	[b]	0.7	<0.7	[b]	<0.7
Penrose	1.8	1.3	1.8	1.0	[b]	1.5	0.7	2.2	[b]	<0.7

[a] Values were given in units dpm g^{-1} in the source documents (CDH 1977 and CDH 1990).
[b] No sample result was available for this location for the indicated year.

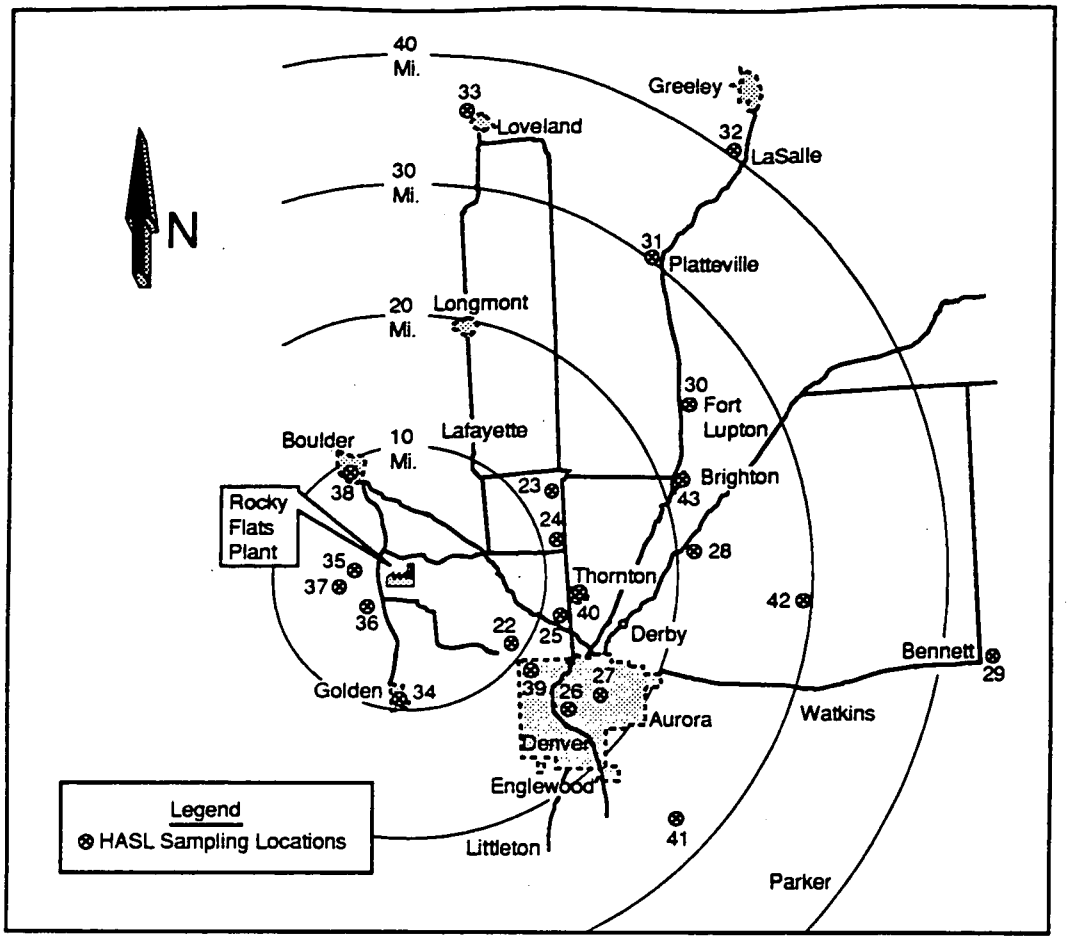


Figure 4. Locations of HASL soil sampling around the Rocky Flats Plant. Locations 1–33 were used in the 1970 sampling (Krey and Hardy 1970 and Seed et al. 1971) and the 1971 sampling (Krey and Krajewski 1972). Locations 34–43 were added in the 1972 sampling (Krey 1976). Only locations numbered higher than 22 are shown here. Other locations are close to the plant.

Krey and Hardy (1970) did not measure or calculate background Pu concentrations in soil from their 1970 sampling. They report a background concentration of 1.5 mCi km^{-2} (56 Bq m^{-2}), based on a single measurement in 1965 in Derby, Colorado (Figure 4).

Seed et al. (1971) performed additional analyses on the data of Krey and Hardy, to estimate the background Pu concentration in soil. Seed et al. plotted the distribution of measured concentrations on log-probability paper. This plot indicated that the distribution appeared to be made up of two separate lognormal distributions; one which represented samples dominated by RFP material, and one which represented samples dominated by worldwide fallout Pu. The data were separated into these two subgroups and replotted. Straight lines (on the log-probability plots) were fitted to the data, to obtain statistics about the distributions. From the fitted lognormal distribution, we determined the background distribution to be represented by a median concentration of 2.3 mCi km^{-2} (85 Bq m^{-2}) and

geometric standard deviation 1.16 (Seed et al. indicated an average value of 2.4 mCi km^{-2} (89 Bq m^{-2}).

Krey and Krajewski (1972) used isotopic ratios to evaluate RFP and fallout contributions to total Pu in soil. In October 1971 they obtained additional soil samples from locations 24 and 28 of the previous HASL sampling by Krey and Hardy (1970) (see Figure 4). The new samples were taken to a depth of 10 cm. The sample analyses were for isotopic ^{239}Pu and ^{240}Pu, in addition to total $^{239,240}Pu$. Ratios of ^{240}Pu to ^{239}Pu were then calculated for the samples at locations 24 and 28, as well as for two "reference" locations known to contain primarily fallout Pu and primarily RFP Pu, respectively. Because the ratios for RFP Pu and worldwide fallout Pu were significantly different, it was possible to calculate, for locations 24 and 28, the amounts of Pu that originated from fallout and from the RFP. The total measured ^{239}Pu concentrations at locations 24 and 28 were 2.39 (± 2.5%) and 1.67 (± 2.5%) mCi km^{-2} (88 and 62 Bq m^{-2}) respectively. For these two locations, the concentrations of ^{239}Pu that originated from fallout were then calculated to be 1.49 and 1.52 mCi km^{-2} (55 and 56 Bq m^{-2}). Thus, Krey and Krajewski estimated the background concentration of Pu due to fallout to be 1.5 mCi km^{-2} (56 Bq m^{-2}).

Krey (1976) applied the isotopic ratio methods of Krey and Krajewski (1972) to an expanded sampling program. In September and October 1972, soil samples were collected from previous locations 22, 23, 27, and 29–32, and from ten new locations, 34–43 (see Figure 4). As seen in the figure, these locations ranged from a few miles from the RFP to about 40 miles from the plant. For this study, sample depth was 10 cm, as that depth was thought to contain about 90% of the deposited Pu. For the analysis, Krey also included the results from locations 24 and 28 from the previous study of Krey and Krajewski (1972). Total measured deposition of $^{239,240}Pu$ was 1.13–2.87 mCi km^{-2} (41.8–106 Bq m^{-2}). From the ratios of ^{240}Pu to ^{239}Pu, the $^{239,240}Pu$ deposition due to the RFP was calculated. We have performed the subtraction to obtain the estimated $^{239,240}Pu$ deposition due to global fallout. The global fallout deposition was 1.12–2.51 mCi km^{-2} (41.4–92.9 Bq m^{-2}). The mean $^{239,240}Pu$ deposition due to global fallout was calculated by Krey to be 1.7 ± 0.5 mCi km^{-2} (63 ± 20 Bq m^{-2}). Table 11 summarizes the estimated background concentrations of Pu in soils, based on the HASL studies.

In summary, the measurements performed by NCAR and CDH (Table 3, Table 4, and Table 5) indicate that the background concentration of $^{239,240}Pu$ in surface soils (0.3 and 1 cm depths) of eastern Colorado is in the range of about 0.3–4.5 Bq kg^{-1} (0.008–0.1 pCi g^{-1}), though only one value was greater than 2.3 Bq kg^{-1}. The wide variability in results may be due to the very shallow surface layers of soil that were sampled, and to the spatial patterns of deposition across the large area covered by sampling. To summarize the measurements and analyses performed by HASL (and the analysis of HASL results by Seed et al. (1971)) we use the values (from Table 6) and one standard deviation or one geometric standard deviation (where available) to represent likely ranges. These results indicate that the total deposition of $^{239,240}Pu$ by the early 1970s from global fallout, in the general area around the RFP, was probably in the range of 40–100 Bq m^{-2} (1.1–2.7 mCi km^{-2}).

Table 11. Summary of Determinations of Background (Global Fallout) Total Deposition of $^{239,240}Pu$ in Soils within 40 Miles of the Rocky Flats Plant, by HASL

Date	Sites	Deposition of $^{239,240}Pu$ [a] ($mCi\ km^{-2}$)	($Bq\ m^{-2}$)	Determination method	Reference
1965	1	1.5	56	"background" location	Krey and Hardy 1970
1970	33	2.3 ×/+ 1.16 [b]	85 ×/+ 1.16 [b]	log-probability analysis of distribution of results	Seed et al. 1971
1971	2	1.50	56	^{240}Pu:^{239}Pu ratios	Krey and Krajewski 1972
1972	19	1.7 ± 0.5 [c]	63 ± 20 [c]	^{240}Pu:^{239}Pu ratios	Krey 1976

[a] Results were reported in source documents in units $mCi\ km^{-2}$.
[b] The ×/+ value here is one geometric standard deviation of the samples.
[c] The ± value here is one standard deviation of the average.

BACKGROUND PLUTONIUM IN SOIL AT GREATER DISTANCES FROM THE ROCKY FLATS PLANT

In this section we describe measurements of background soil concentrations of Pu for locations farther from the RFP. While concentrations at great distances from the plant may not be indicative of the background around the plant, they do provide some perspective as to how local background concentrations compare with regional and global background.

Purtymon et al. (1990) reports data on soil concentrations of Pu in northern New Mexico and southern Colorado, which are in the same general region as the RFP. In this study, six locations were sampled in 1981 and 1983, and nine separate locations were sampled in 1986. The locations were all east of (or on) the continental divide. The northern-most location was Monarch Pass, Colorado, about 100 miles from the RFP, and the southern-most location was Santa Ana Pueblo, New Mexico, about 300 miles from the RFP. Some of the locations are, however, within about 20 miles of the Los Alamos National Laboratory, which was a potential source of Pu in the environment. The soil samples were composites made up of five sub-samples, taken to depth 5 cm. Alpha spectroscopy measurements were performed to obtain ^{238}Pu and $^{239,240}Pu$, which were summed to obtain total Pu. On the average, ^{238}Pu contributed less than 5% to the total Pu activity measured. We only consider the $^{239,240}Pu$ measurements here, for comparability with other measurements. Concentrations of $^{239,240}Pu$ ranged from 1.2 to 81.0 $fCi\ g^{-1}$ (0.044 to 3.00 $Bq\ kg^{-1}$), with average 14.4 $fCi\ g^{-1}$ (0.533 $Bq\ kg^{-1}$) and standard deviation 17.7 $fCi\ g^{-1}$ (0.655 $Bq\ kg^{-1}$). The two highest values occurred for locations in high mountain passes on the continental divide. As discussed earlier, higher values are expected for high mountain areas, and it may be reasonable to consider these locations grouped separately from the remaining locations. If these highest values are disregarded, the remainder cover the range 1.2–19.3 $fCi\ g^{-1}$ (0.044–0.714 $Bq\ kg^{-1}$), with average 9.4 $fCi\ g^{-1}$ (0.35 $Bq\ kg^{-1}$) and standard deviation 5.4 $fCi\ g^{-1}$ (0.20 $Bq\ kg^{-1}$).

As mentioned above, Holleman et al. (1987) provides an extensive compilation of data sets on worldwide fallout of Pu from weapons tests. From this compilation, we extracted measured concentrations of $^{239,240}Pu$ in soil in the U.S. Values were given in units of total deposition (Bq m^{-2}) and mass concentration (Bq kg^{-1}). We use the same units. Information about individual measurements is given in Table 14, at the end of this memorandum. Holleman et al. does not provide information about sample depths, but this is not necessary for our work. The results are summarized by state in Table 12. We note that the samples in Ohio were taken near the Mound facility, which processed Pu, thought this facility handled primarily ^{238}Pu. The very low values of deposition reported for Alaska are probably due to the more northerly latitude of Alaska. The low deposition value for Colorado (2.11 Bq m^{-2}) does not appear to be a credible value; deposition this small seems extremely unlikely.

Table 12. Summary of $^{239,240}Pu$ Deposition and Mass Concentrations in Soil in the United States, from the Compilation by Holleman et al. (1987)

				Deposition (Bq m^{-2}) [a]			Concentration (Bq kg^{-1}) [a]		
State	Sites	Samples	Dates	Min	Max	Avg	Min	Max	Avg
Alaska	5	9	1964–1976	1.18	34	13			
California	3	5	1970–1972	27	37	32			
Colorado	1	7	1965–1970	2.11	67	47			
Florida	1	1	1970			37			
Hawaii	1	1	1970			148			
Illinois	17	62	1970–1981	10.36	256.78	51			
Kansas	1	1	1970			89			
Maine	1	1	1970			63			
Massachusetts	1	1	1972			85			
Michigan	1	1	1976			99.9			
Montana	1	1	1965			70			
New Mexico	6	36	1974–1977				0.0	0.78	0.28
New York	7	16	1964–1973	67	99.9	84			
North Carolina	1	1	1970			89			
Ohio	1	25	1974				0.114	1.528	0.28
Oklahoma	1	1	1970			81			
South Dakota	2	2	1965–1970	85	93	89			
Texas	2	2	1970	32.6	36.6	35			
Utah	1	1	1970			96			
Washington	2	7	1970–1971	1.5	52	20			
Wisconsin	1	1	1972			58			

a "Min" means minimum, "max" means maximum, and "avg" means arithmetic average. Minimum and maximum values are taken from the source document (Holleman et al. 1987), and the averages are calculated by us, in this present work. The values presented retain the significant figure used by Holleman et al., though we acknowledge that in some cases they are excessive.

CONCLUSIONS ON BACKGROUND PLUTONIUM IN SOILS AROUND THE RFP

The measured levels of Pu in soil around the RFP and around the United States, from the studies we have presented here, are compared in Table 13. (We acknowledge that this is

not necessarily a complete compilation of such data.) The measured deposition concentrations of $^{239,240}Pu$ around the RFP (40–100 Bq m^{-2}, or 1.1–2.7 mCi km^{-2}) are within the ranges seen for other states (1.2–260 Bq m^{-2}, or 0.03–7 mCi km^{-2}), though they tend toward the higher end of measured concentrations. The maximum measured mass concentrations of $^{239,240}Pu$ around the RFP (<0.3–4.5 Bq kg^{-1}, or 0.008–0.1 pCi g^{-1}) exceed (slightly) the ranges of values seen in New Mexico and Ohio measurements (0–3.0 Bq kg^{-1}, or 0–0.08 pCi g^{-1}). Many of the lowest values for the United States are probably for locations, such as Alaska, not in the middle latitudes. Thus, it appears that measured background concentrations of Pu in soil around the RFP tend to be higher than background concentrations for many locations, but are still within the ranges observed in other states.

Table 13. Comparison of Measured Deposition and Mass Concentrations of $^{239,240}Pu$ Around the Rocky Flats Plant, with those Around the United States

	Deposition (Bq m^{-2})		Concentration (Bq kg^{-1})	
Locations	Minimum	Maximum	Minimum	Maximum
Around RFP	40	100	<0.3	4.5
United States [a]	1.2	260	0.0	3.0

[a] Mass concentrations were from two states only: New Mexico and Ohio.

There are some important characteristics related to Pu in soils that should be considered in evaluations of soil sample results around the RFP. The measurement technique, and more specifically the Pu isotopes actually measured should be determined. Most measurements of "^{239}Pu" are actually measurements of $^{239,240}Pu$, because alpha spectroscopy is commonly used for the analyses. If isotopic results, such as the ratio $^{240}Pu/^{239}Pu$ are available, it may be feasible to determine more accurately whether the source of the Pu is truly background, or has been influenced by RFP sources. When comparing samples near the RFP to background samples, the time of sample collection can be important, as there are temporal trends in the global fallout of Pu from nuclear weapons testing. Finally, depth distributions of Pu should be considered relative to the goals of a particular sampling program or analysis. Soil samples taken from the surface soils (e.g. to 1 cm or so) are not representative of the total deposition of Pu that exists in the soil column. Strict quantitative comparisons between results of sampling programs with widely disparate sample depths may be difficult or unwise.

REFERENCES

Bennett B.G. 1978. *Environmental Aspects of Americium*. Rep. EML-348, Environmental Measurements Laboratory, U.S. Department of Energy, New York, New York.

CCEI (Colorado Committee for Environmental Information). 1970. *Report on the Dow Rocky Flats Fire: Implications of Plutonium Releases to the Public Health and Safety*. Report dated January 13, 1970, with attached addendum of more recent results. CCEI, Subcommittee on Rocky Flats, Boulder, Colorado.

CDH (Colorado Department of Health). 1977. *Radioactive Soil Contamination (Cesium-137 and Plutonium) in the Environment near the Rocky Flats Nuclear Weapons Plant.*

CDH (Colorado Department of Health). 1990. *Rocky Flats Surface Soil Survey 1970 - 1989.* Radiation Control Division, CDH.

Eisenbud M. 1987. *Environmental Radioactivity: from Natural, Industrial, and Military Sources.* Academic Press, Inc., San Diego, California.

Harley J.H. 1979. *Plutonium in the Environment - a Review.* Paper prepared for the 1979 annual meeting of the Radiation Research Society of Japan. Environmental Measurements Laboratory, U.S. Department of Energy, New York, New York.

Holleman J.W., P.A. Quiggins, B.D. Chilton, M.S. Uziel, H.A. Pfuderer, and J.A. Longmire. 1987. *Worldwide Fallout of Plutonium from Nuclear Weapons Tests.* Rep. ORNL-6315, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Krey P.W. 1976. "Remote Plutonium Contamination and Total Inventories from Rocky Flats." *Health Physics* **30**:209–214.

Krey P.W. and E.P. Hardy. 1970. *Plutonium in Soil around the Rocky Flats Plant.* Rep. HASL-235, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York, New York.

Krey P.W. and B.T. Krajewski. 1972. "Plutonium Isotopic Ratios at Rocky Flats." In: Hardy E.P. *Health and Safety Laboratory, Fallout Program Quarterly Summary Report (December 1, 1971 through March 1, 1972).* Rep. HASL-249, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York, New York.

Little C.A. and F.W. Whicker. 1978. "Plutonium Distribution in Rocky Flats Soil." *Health Physics* **34**:451–457.

Poet S.E. and E.A. Martell. 1972. "Plutonium-239 and Americium-241 Contamination in the Denver Area." *Health Physics* **23**:537–548.

Purtymon W.D., R.J. Peters, and M.N. Maes. 1990. *Plutonium Deposition and Distribution from Worldwide Fallout in Northern New Mexico and Southern Colorado.* Rep. LA-11794-MS, Los Alamos National Laboratory, Los Alamos, New Mexico.

Seed J.R., K.W. Calkins, C.T. Illsley, F.J. Miner, and J.B. Owen. 1971. *Committee Evaluation of Plutonium Levels in Soil within and Surrounding USAEC Installation at Rocky Flats, Colorado.* Rep. RFP-INV-10, Rocky Flats Division, The Dow Chemical Company, Golden, Colorado.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). 1993. *Sources and Effects of Ionizing Radiation.* United Nations, New York, New York.

Webb S.B., S.A. Ibrahim, and F.W. Whicker. 1993. "A Study of Plutonium in Soil and Vegetation at the Rocky Flats Plant." In: Kathren R.L., D.H. Denham, and K. Salmon. 1993. *Environmental Health Physics: Proceedings of the Twenty-Sixth Midyear Topical Meeting of the Health Physics Society, January 24–28, 1993, Coeur d'Alene, Idaho.* Columbia Chapter, Health Physics Society, Richland, Washington.

Whicker F.W., C.A. Little, and T.F. Winsor. 1974. "Plutonium Behaviour in the Terrestrial Environs of the Rocky Flats Installation." In: International Atomic Energy Agency (IAEA). *Environmental Surveillance Around Nuclear Installations.* Vol. 2, pages 89–103. Rep. IAEA-SM-180/45, IAEA, Vienna, Austria.

ANNEX

Table 14. Individual Measurements of $^{239,240}Pu$ in Soil in the United States, from the Compilation of Holleman et al. (1987)

State	Location	Date	$^{239,240}Pu$ concentration	
			Bq m^{-2}	Bq kg^{-1}
Alaska	Anaktuvuk Pass	Jul 1975	5.62	
Alaska	Anaktuvuk Pass	Jul 1976	1.55	
Alaska	Anaktuvuk Pass	Sep 1976	1.18	
Alaska	Barrow	Aug 1964	12.20	
Alaska	Barrow	1970	14.8	
Alaska	Bettles	Jul 1976	4.26	
Alaska	Fairbanks	Jul 1976	8.21	
Alaska	Fairbanks	1970	31.4	
Alaska	Palmer	1970	34	
California	Burbank	1970	27	
California	Oakland	Oct 1972	30.00	
California	Oakland	Oct 1972	30.00	
California	San Francisco	Oct 1972	34.00	
California	San Francisco	Oct 1972	37.00	
Colorado	Denver	Sep 1965	56	
Colorado	Denver	Feb 1970	32.9	
Colorado	Denver	Feb 1970	40.7	
Colorado	Denver	Sep 1970	2.11	
Colorado	Denver	Sep 1970	65	
Colorado	Denver	Oct 1970	67	
Colorado	Denver	1970	67	
Florida	Ft. Pierce	1970	37	
Hawaii	Papaikou	1970	148	
Illinois	Argonne	1970	78	
Illinois	Brookfield	Sep 1972	57.35	
Illinois	Brookfield	Oct 1974	65.86	
Illinois	Brookfield	Jun 1976	70.3	
Illinois	Brookfield	Jun 1979	36.63	
Illinois	Brookfield	Oct 1980	49.21	
Illinois	Channahon	Jun 1978	49.6	
Illinois	Channahon	Jun 1979	31.08	
Illinois	Channahon	Jun 1980	19.61	
Illinois	Channahon	Jun 1981	25.2	
Illinois	Downers Grove	Jun 1979	18.5	
Illinois	Downers Grove	Oct 1981	29.2	
Illinois	Dresden Lock and Dam	Oct 1976	74	
Illinois	Dresden Lock and Dam	Oct 1978	45.1	
Illinois	Dresden Lock and Dam	Oct 1979	15.91	
Illinois	Dresden Lock and Dam	Oct 1980	10.36	
Illinois	Dresden Lock and Dam	Oct 1981	41.8	
Illinois	Hinsdale	Oct 1974	127.65	
Illinois	Hinsdale	Jun 1976	81.4	
Illinois	Lemont	Nov 1974	56.61	
Illinois	Lemont	Oct 1978	21.5	

Table 14. Individual Measurements of $^{239,240}Pu$ in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

State	Location	Date	$^{239,240}Pu$ concentration	
			Bq m^{-2}	Bq kg^{-1}
Illinois	Lemont	Jun 1980	19.61	
Illinois	Lemont	Oct 1981	23.7	
Illinois	McGinnis Slough	Sep 1972	72.52	
Illinois	McGinnis Slough	May 1974	80.3	
Illinois	McGinnis Slough	Oct 1978	42.5	
Illinois	McGinnis Slough	Jun 1980	22.57	
Illinois	Mckinley Woods State Park	Jun 1972	40.7	
Illinois	Mckinley Woods State Park	Oct 1974	77.7	
Illinois	Mckinley Woods State Park	Jun 1976	114.7	
Illinois	Mckinley Woods State Park	Oct 1978	54.8	
Illinois	Mckinley Woods State Park	Oct 1979	35.52	
Illinois	Mckinley Woods State Park	Oct 1980	20	
Illinois	Mckinley Woods State Park	Oct 1981	69.2	
Illinois	Morris	May 1974	75.85	
Illinois	Morris	May 1974	256.78	
Illinois	Morris	Jun 1978	52.2	
Illinois	Morris	Jun 1979	32.56	
Illinois	Morris	Jun 1980	27	
Illinois	Morris	Jun 1981	17	
Illinois	Naperville	Jun 1972	55.5	
Illinois	Naperville	May 1974	94	
Illinois	Naperville	Jun 1978	57.7	
Illinois	Naperville	Jun 1981	24	
Illinois	Romeoville	Oct 1978	57.3	
Illinois	Romeoville	Oct 1981	44.4	
Illinois	Saganashkee Slough	Jun 1972	77.33	
Illinois	Saganashkee Slough	May 1974	72.52	
Illinois	Saganashkee Slough	Oct 1978	27	
Illinois	Saganashkee Slough	Jun 1980	21.83	
Illinois	Starved Rock State Park	May 1974	76.22	
Illinois	Starved Rock State Park	Jun 1978	43.3	
Illinois	Starved Rock State Park	Jun 1979	31.08	
Illinois	Starved Rock State Park	Jun 1980	17.39	
Illinois	Starved Rock State Park	Jun 1981	43.7	
Illinois	Western Springs	Jun 1979	35.9	
Illinois	Western Springs	Oct 1980	24.8	
Illinois	Willow Springs	Oct 1976	107.3	
Illinois	Willow Springs	Jun 1978	27	
Illinois	Willow Springs	Oct 1979	30.71	
Illinois	Woodridge	Oct 1979	32.56	
Illinois	Woodridge	Jun 1981	30.3	
Kansas	Manhattan	1970	89	
Maine	Orono	1970	63	
Massachusetts	North Eastham, Cape Cod	Oct 1972	85	
Michigan	St. Joseph	Oct 1976	99.9	
Montana	Bozeman	Sep 1965	70	

Table 14. Individual Measurements of $^{239,240}Pu$ in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

State	Location	Date	$^{239,240}Pu$ concentration	
			Bq m^{-2}	Bq kg^{-1}
New Mexico	Bernalillo	Jul 1974		0.22
New Mexico	Bernalillo	May 1975		0.44
New Mexico	Bernalillo	Oct 1975		0.04
New Mexico	Bernalillo	Apr 1976		0.15
New Mexico	Bernalillo	Oct 1976		0.07
New Mexico	Bernalillo	Mar 1977		0
New Mexico	Bernalillo	Oct 1977		0.07
New Mexico	Chamita	Jul 1974		0.22
New Mexico	Chamita	Oct 1975		0.63
New Mexico	Chamita	Mar 1976		0.3
New Mexico	Chamita	Oct 1976		0.52
New Mexico	Chamita	Mar 1977		0.63
New Mexico	Chamita	Oct 1977		0.37
New Mexico	Cochiti	May 1975		0.07
New Mexico	Cochiti	Oct 1975		0
New Mexico	Cochiti	Apr 1976		0.15
New Mexico	Cochiti	Oct 1976		0.11
New Mexico	Cochiti	Mar 1977		0.04
New Mexico	Cochiti	Oct 1977		0.11
New Mexico	Embudo	Jul 1974		0.19
New Mexico	Embudo	May 1975		0.3
New Mexico	Embudo	Oct 1975		0.33
New Mexico	Embudo	Mar 1976		0.44
New Mexico	Embudo	Oct 1976		0.7
New Mexico	Embudo	Mar 1977		0.4
New Mexico	Embudo	Oct 1977		0.56
New Mexico	Jemez	Jul 1974		0.04
New Mexico	Jemez	May 1975		0.04
New Mexico	Jemez	Sep 1975		0.44
New Mexico	Jemez	Apr 1976		0.07
New Mexico	Jemez	Oct 1976		0.26
New Mexico	Jemez	Mar 1977		0.7
New Mexico	Jemez	Oct 1977		0.04
New Mexico	Otowi	Jul 1974		0.44
New Mexico	Otowi	May 1975		0.22
New Mexico	Otowi	Oct 1977		0.78
New York	Bronx	Jul 1970	92.5	
New York	Brookhaven National Laboratory	Sep 1970	78	
New York	Brookhaven National Laboratory	Sep 1970	96	
New York	Brookhaven National Laboratory	Nov 1972	81	
New York	Brookhaven National Laboratory	Nov 1972	90.6	
New York	Brookhaven National Laboratory	Nov 1972	91.8	
New York	Brookhaven National Laboratory	Nov 1972	99.9	
New York	Brookhaven National Laboratory	1972	88.8	
New York	Brooklyn	Nov 1972	78	
New York	Fordham University	Dec 1969	74	

Table 14. Individual Measurements of $^{239,240}Pu$ in Soil in the United States, from the Compilation of Holleman et al. (1987) (continued)

State	Location	Date	$^{239,240}Pu$ concentration	
			Bq m^{-2}	Bq kg^{-1}
New York	Fordham University	Jan 1970	81.4	
New York	Fordham University	Jan 1970	96	
New York	Kitchawan, Westchester County	Jun 1973	70.3	
New York	New York	Dec 1964	67	
New York	New York	1970	96	
New York	Teatown, Westchester County	Jun 1973	70.3	
North Carolina	Raleigh	1970	89	
Ohio	Miamisburg	Oct 1974		0.114
Ohio	Miamisburg	Oct 1974		0.129
Ohio	Miamisburg	Oct 1974		0.135
Ohio	Miamisburg	Oct 1974		0.14
Ohio	Miamisburg	Oct 1974		0.16
Ohio	Miamisburg	Oct 1974		0.166
Ohio	Miamisburg	Oct 1974		0.17
Ohio	Miamisburg	Oct 1974		0.171
Ohio	Miamisburg	Oct 1974		0.171
Ohio	Miamisburg	Oct 1974		0.174
Ohio	Miamisburg	Oct 1974		0.177
Ohio	Miamisburg	Oct 1974		0.179
Ohio	Miamisburg	Oct 1974		0.18
Ohio	Miamisburg	Oct 1974		0.19
Ohio	Miamisburg	Oct 1974		0.191
Ohio	Miamisburg	Oct 1974		0.2
Ohio	Miamisburg	Oct 1974		0.206
Ohio	Miamisburg	Oct 1974		0.207
Ohio	Miamisburg	Oct 1974		0.208
Ohio	Miamisburg	Oct 1974		0.213
Ohio	Miamisburg	Oct 1974		0.222
Ohio	Miamisburg	Oct 1974		0.256
Ohio	Miamisburg	Oct 1974		0.269
Ohio	Miamisburg	Oct 1974		1.214
Ohio	Miamisburg	Oct 1974		1.528
Oklahoma	Tulsa	1970	81	
South Dakota	Rapid City	Sep 1965	93	
South Dakota	Vermillion	1970	85	
Texas	Kingsville	1970	36.6	
Texas	Weslaco	1970	32.6	
Utah	Salt Lake City	1970	96	
Washington	Hanford Reservation	Feb 1971	1.5	
Washington	Hanford Reservation	Feb 1971	7.8	
Washington	Hanford Reservation	Feb 1971	8.1	
Washington	Hanford Reservation	Feb 1971	19.20	
Washington	Hanford Reservation	Feb 1971	24.00	
Washington	Hanford Reservation	Feb 1971	28.10	
Washington	Puyallup	1970	52	
Wisconsin	Lake Delavan	Oct 1972	58.46	